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(54) Title: IMPROVED PRESSURE SENSITIVE LABELING ADHESIVE (57) Abstract A pressure sensitive adhesive which exhibits both improved adhesive characteristics and improved convertibility. This adhesive is obtained by combining a typical styrenic block copolymer, blended with tackifying resins, whether natural or hydrocarbon based, and including, a styrene isoprene di-block copolymer. The effect of this use of styrene isoprene di-block copolymer, besides the expected result of reducing the tendency of conventional plasticizers to migrate from the adhesive and cause staining of the label stock or release liner, also has the unexpected results of substantially improved temperature performance and a very substantial improvement in ability to be die cut.		

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IMPROVED PRESSURE SENSITIVE LABELING ADHESIVE

Technical Field

This invention relates to novel pressure sensitive adhesive compositions, and in particular to such pressure sensitive adhesives which are best suited to the manufacture of pressure sensitive labels.

Background of the Invention

Pressure sensitive adhesives are materials which have tack properties at room temperature. The pressure sensitive adhesive firmly adheres to a variety of dissimilar surfaces without the need of more than finger or hand pressure.

During label manufacture a laminate, formed of a face stock, a pressure sensitive adhesive layer, and a release liner, is passed through apparatus which converts the laminate to yield commercially useful labels and label stock. The processes involved in the converting operation include, for example, printing, die cutting and matrix stripping to leave labels on a release liner, butt cutting of labels to the release liner, marginal hole punching, perforating, fan folding, guillotining and the like. Die and butt cutting involve cutting of the laminate to the face of the release liner. Other procedures involve cutting entirely through the label laminate and include, for instance, hole punching and perforating, and guillotining.

The cost of converting a laminate into a finished product is a function of the speed at which the various processing operations can be done. While the nature of all layers of the laminate can impact cost of convertibility, historically the adhesive layer has been the greatest limiting factor in the ease and cost of the conversion operation. This is true because of the viscoelastic nature of the adhesive. The fact that the adhesive is viscoelastic hampers precise and clean penetration of a die in die cutting operations, and promotes adherence to cutting blades and the like in any type of cutting

operation. The fact that adhesives can be stringy also has an impact on matrix stripping operations, which are often done after die cutting operations.

To date, achieving good convertibility has not automatically coincided with achieving excellent adhesive performance. Adhesives must be formulated to fit predetermined needs. Important properties include peel adhesion, tack, shear properties, viscosity at various temperatures, and the like. Good general purpose adhesives may exhibit poor convertibility simply because the adhesive is difficult to cleanly cut. Such an adhesive may stick to a die or blade. In label manufacture, die cutting and matrix stripping operations by necessity occur at a variety of speeds ranging up to 1000 feet per minute or more. Within that range, an adhesive may provide regions where a matrix will break despite the fact that successful matrix stripping can occur at speeds on either side of the region. It is a goal to provide adhesive systems wherein the adhesive can be cleanly cut and the matrix stripped at substantially any practical operating speed.

Sasaki et al, U.S. Patent No. 5,290,842 shows a combination wherein a styrene-butadiene block copolymer is combined with a styrene-isoprene-styrene block copolymer to show two separate and distinct glass transition temperature peaks, allegedly to improve the convertibility. Other patents of interest are Downey, U.S. Patent No. 3,880,953; Tindall, U.S. Patent No. 3,509,239; Feeney, U.S. Patent No. 4,060,503; and Korpman, U.S. Patent No. 3,932,328.

This invention relates to improvements to the products described above and to solutions to some of the problems raised thereby.

Summary of the Invention

This invention provides a pressure sensitive adhesive which exhibits improved convertibility, that is, the ability to achieve cutting of the adhesive in processing operations involving cutting through a face stock and adhesive to at least the release liner of the

laminate, while at the same time providing superior adhesive performance.

These results can be obtained by combining a typical styrenic block copolymer, blended with tackifying resins, whether natural or hydrocarbon based, and including, in place of some or all of the plasticizer, a styrene isoprene di-block copolymer. Both the styrenic block copolymer and the styrene isoprene di-block copolymer are miscible together with the tackifier to produce a pressure sensitive adhesive that exhibits a single glass transition temperature (T_g) value. The expected result of this partial or complete replacement of the plasticizer is mainly the reduction of the tendency of the plasticizer to migrate from the adhesive and cause staining of the face stock, the release liner, or both. Also, however, this partial or complete replacement of the plasticizer produces the unexpected results of substantially higher temperature performance and a very substantial improvement in the ability of the adhesive to be die cut.

Previous attempts to improve the die cutting of hot melt pressure sensitive adhesives have had detrimental effects on other characteristics. For example, increasing the resin content increases the tangent delta value of the pressure sensitive adhesive compared to the tangent delta value measured in the absence of increased resin content, thereby increasing the ease of die cutting, but also significantly increases the glass transition temperature, T_g . The T_g can be increased to a point where the adhesive no longer maintains its pressure sensitive characteristics. In addition, the application and service temperature ranges are condensed. Incorporation of or increasing the level of typical processing oils will improve the processability, but will decrease an adhesive's ability to be die cut, and will diminish high temperature performance. The addition of a low molecular weight styrene-isoprene di-block copolymer improves processability and improves the tangent delta values at 20°C, while maintaining an acceptably low T_g . This enhancement in ability to be die cut and the

enhancement in high temperature performance are unexpected benefits. A preferred plasticizer is composed of a styrene isoprene di-block copolymer and a processing oil in a weight ratio of about 1:2 to 1:0.5.

- 5 Other objects and advantages of the invention will become apparent hereinafter.

Detailed Description of the Invention

The present invention is directed in general to a pressure sensitive adhesive formed from a mixture of base
10 polymers, in particular styrenic block copolymers, and a tackifier. As is generally conventional, the styrenic block copolymers may be selected from any of several types, including but not limited to styrene-isoprene-styrene block copolymers such as Vector 4111 available from Dexco,
15 styrene-isoprene-styrene/styrene-isoprene block copolymers such as Kraton D1107 or Kraton D1112, manufactured and sold by Shell, styrene-butadiene-styrene block copolymers such as Vector 6241D available from Dexco, styrene-butadiene-styrene/styrene-butadiene block copolymers such as Kraton
20 D1102 also from Shell, and styrene-butadiene block copolymers such as Solprene 1205, manufactured and sold by Housmex, Inc. The above examples are unsaturated midblock copolymers. Saturated midblock copolymers are also usable, including but not limited to styrene-ethylene-butylene-
25 styrene block copolymers such as Kraton G1652, available from Shell, styrene-ethylene-butylene-styrene/styrene-ethylene-butylene block copolymers such as Kraton G1657, also from Shell and styrene-ethylene-propylene--styrene/styrene-ethylene-propylene block copolymers such as
30 Septon 2063, manufactured by Kuraray, a Japanese company, and distributed in the United States by Arakawa Chemical Ind., Ltd.

Generally conventional tackifier systems can include, as one example of many choices available, a
35 normally solid tackifier such as Wingtack 95, an aliphatic petroleum resin, manufactured by Goodyear, or Escorez 1310 LC, an aliphatic petroleum resin, manufactured by Exxon,

together with a plasticizer system, usually including a plasticizer oil such as Shellflex 371, from Shell, or a normally liquid tackifier such as Wingtack 10, an aliphatic petroleum resin, available from Goodyear, or some
5 combination of both.

The invention calls for the addition of a styrene isoprene di-block copolymer having a low viscosity, preferably a viscosity such that the styrene isoprene di-block copolymer is a liquid at 25°C. This styrene isoprene
10 di-block copolymer used in the adhesive composition of the present invention has the configuration of A-B, wherein "A" is a styrene polymer block and "B" is an isoprene polymer block. These "di-block" copolymers used in the present invention differ significantly from "tri-block" polymers
15 having an A-B-A configuration, with "A" and "B" each representing separate, distinct polymers. Di-block copolymers are also distinguished from "radial polymers" (AB_x) which consist of a central "A" block of a polymer with numerous arms of a "B" block of a different polymer
20 extending from the central A polymer block. The preferred styrene isoprene di-block copolymers of the present invention have an absolute molecular weight of less than 70,000, with the most preferred styrene isoprene di-block copolymers having an absolute molecular weight of less than
25 50,000, or a polystyrene equivalent molecular weight of less than 70,000. Preferred A-B styrene isoprene di-block copolymers of the present invention will have the styrene A block amounting to about 5% to about 50% of the mass of the copolymer molecule, most preferably about 5% to about 25%.
30 In particular, LVSI-101 is one such styrene isoprene di-block copolymer available from Shell, and LIR-310 is another, available from Kuraray through Arakawa, for this purpose. The plasticizers referred to above, and particularly the oils, have tended to be subject to
35 migration, causing adverse effects on substrate and adhesive performance. Since styrene isoprene di-block copolymer is substantially less migratory than these plasticizers, the addition of the styrene isoprene di-block

copolymer in place of some or all of the normally liquid tackifier and/or plasticizer oil was expected to have the effect of reducing staining. What was not expected, however, was that the inclusion of the styrene isoprene di-
5 block copolymer provided broad end-use temperature ranges while at the same time substantially improving die-cutting performance.

In order to demonstrate and verify these effects, several formulations were prepared, using generally
10 standard preparation processes and methods, according to the Examples 1 through 4 shown in the following tables, and certain testing procedures were employed.

Dynamic Mechanical Analysis, or DMA, is used to measure how a material responds to an imposed strain or
15 deformation. This response will either be a viscous, elastic or viscoelastic response. DMA is used to model and predict how a material will respond to real-world phenomena such as coating, die-cutting, aging and other conditions. DMA is also used to predict adhesive performance. The DMA
20 testing used to profile these pressure-sensitive adhesives was run on 25 mm parallel plates using a frequency of 10 radians per second and a 1 or 5 percent strain, as indicated. Temperature sweeps were performed from 140° to -40°C with an initial gap of 1.6 mm.

25 The "tangent delta" is the ratio of the viscous response (G'') to the elastic response (G') of a particular DMA curve. The glass transition temperature (T_g) is indicated by a peak on the tangent delta curve of a temperature sweep. Die-cutting performance for the
30 formulations referred to herein can be approximated by comparing the tangent delta values at 20°C for the different formulations. A higher tangent delta value at a given temperature indicates a material will respond in a more viscous fashion. The greater the viscous response,
35 the more likely a material will flow when subjected to an outside force. Consequently, materials with higher tangent delta values at a temperature of interest will tend to die-cut better. They flow and separate when subjected to

cutting forces, as compared to materials with low tangent delta values which deform and then recover. Hence, the higher the tangent delta value at 20°C, the better the die-cutting performance. The tangent delta value and the T_g of a formulation, along with various performance tests, will determine the suitability of a product for a pressure-sensitive application. The addition of the styrene isoprene di-block copolymer in the present invention results in a pressure sensitive adhesive having an increased tangent delta value of 5% or greater when compared to a similar pressure sensitive adhesive which is identical except for lacking the addition of the styrene isoprene di-block copolymer.

The pressure sensitive adhesive compositions of the present invention may be formulated using any of the techniques well known in the art. A representative example of a prior art procedure involves placing all of the oil substances and any optional stabilizer substances in a jacketed mixing kettle, and preferably in a jacketed heavy duty mixer of the Baker-Perkins or Day type, equipped with rotors. The materials are mixed and the temperature of the mixture is raised to a range from about 250°F to about 350°F. As should be understood, the precise temperature to be used in this step will depend on the melting point of the particular ingredients. When the initial mixture has been heated, the mixture is blanketed with CO_2 at a slow flow rate and the required tackifiers, or resins, described above, are slowly added. When all of the ingredients are melted at the desired temperature, the copolymers are added to the mixture. The resultant pressure sensitive adhesive composition mixture is agitated until the copolymers are completely dissolved. A vacuum is then applied to remove any entrapped air.

The preparation of coated stock for pressure sensitive adhesive testing is described as follows. Adhesive is coated onto a release liner (SBL 42 SC FLU from Akrosil or equivalent) such that a 2.25 inch wide continuous pattern is applied at 1.0 ± 0.05 mils (adhesive

dry weight). A two mil polyester film is laminated to the adhesive within 5 to 5.5 seconds (web speed of approximately 10 ft./min.) for use in 180° peel and SAFT testing.

- 5 One type of testing, 180° peel testing, is designed to measure the strength of an adhesive bond to a standard 2 inch by 5 inch stainless steel surface. The strength of the bond is determined by measuring the force required to peel a strip of adhesive-coated polyester stock
10 from a test panel at an angle of 180° after a specified dwell time. The stock is conditioned in a controlled environment ($72 \pm 2^\circ\text{F}/50 \pm 3\%$ relative humidity) for a minimum of 12 hours before 1 inch by 10 inch strips are cut from it. Each strip is applied to the required stainless steel
15 panel, adhesive side down, with a mechanical roll-down machine that uses two passes of a 4.5 pound rubber roller moving at 12 in./min. Each sample is allowed to dwell for 15 minutes before separating at a rate of 12 in./min. on a tensile tester. The peel angle must be maintained at 180°.
20 The resulting peel force is measured and reported in pounds per linear inch. A minimum of three trials should be run and then the average taken.

- Another type of testing is the Shear Adhesion Failure Temperature (SAFT) test. This is a measure of the
25 internal strength of an adhesive at elevated temperatures. An adhesive-coated polyester stock sample is applied to a standard 2.5 inch by 2.5 inch stainless steel panel. A constant load is applied and the temperature raised 3°F per minute until the sample fails from the panel. The stock is
30 conditioned in a controlled environment ($72 \pm 2^\circ\text{F}/50 \pm 3\%$ relative humidity) for a minimum of 12 hours before 1 inch by 3 inch strips are cut from it. Each strip is applied to the required stainless steel panel using sufficient pressure such that a cohesive failure results when the
35 sample fails. Each sample is allowed to dwell for one hour before placing in an oven capable of raising the temperature at a constant rate of 3°F per minute. The test panels are hung vertically in the oven. A 1000 gram weight

(unless otherwise specified) is attached to each sample and allowed to hang until the sample fails. The oven must be conditioned to a constant temperature of 90°F and held for one minute before raising the temperature. The test is run until all samples fail from their respective panels. The temperature at which failure occurs is then recorded. All failures should be cohesive in nature. A cohesive failure results when there is residual adhesive on both the test panel surface and the coated substrate. An adhesive failure, indicating delamination from either the coated substrate or the test panel, with no residue, will not result in a representative value. It indicates the temperature at which the adhesive failed from the test panel or substrate, and is no indication of the internal strength of an adhesive. A minimum of three trials should be run and then the average taken. The pressure sensitive adhesive of the present invention preferably demonstrates a rise or a minimal decrease in SAFT. This preferable minimal decrease corresponds to a decrease of 5°F or less for a pressure sensitive adhesive of the instant invention, when compared to a similar pressure sensitive adhesive which is identical except for lacking the addition of the styrene isoprene di-block copolymer.

As to each example formulation, 180° peel tests and shear adhesion failure temperature tests were conducted. The results of those tests on the improved adhesives provided by this invention were compared with the results of those tests conducted on prior art adhesives as controls, and the results tabulated. The glass transition temperature T_g and tangent delta were also calculated and are presented in the same tables.

The pressure sensitive adhesives of the present invention preferably demonstrate a minimal increase in T_g . This minimal increase in T_g corresponds to an increase of 3°C or less for a preferred pressure sensitive adhesive of the instant invention, when compared to a similar pressure sensitive adhesive which is identical except for lacking the addition of the styrene isoprene di-block copolymer.

Table 1 shows controls where, besides the use of Kraton 1107 as the base polymer, and Piccotac 95, from Hercules, as a normally solid tackifier, the mixture includes 15% by weight of conventional plasticizers.

5 Shellflex 371, a hydrocarbon processing oil, is used as the conventional plasticizer in Control 1, while Wingtack 10, a normally liquid tackifier, is used as the conventional plasticizer in Control 2. In Example 1, LVSI-101 is the only ingredient used as a plasticizer. Example 2 includes

10 use of a combination of two-thirds LVSI-101 and one-third Shellflex 371 as a plasticizer, while Example 3 includes use of a combination of one-third LVSI-101 and two-thirds Shellflex 371 as a plasticizer. In each of these formulations Irganox 1010, a phenolic antioxidant, is added

15 to reduce oxidative degradation during processing and to improve shelf life performance. The DMA testing for each of the formulations shown in Table 1 was performed at 5% strain. As can be seen below, each of the examples has peel adhesion, equal or superior to Control 2, which has

20 the liquid tackifier as plasticizer, and clearly superior to Control 1, which has the processing oil as the plasticizer. Further, as indicated by the Shear Adhesion Failure Temperature figures, the examples show significantly increased high temperature strength over

25 conventional adhesives.

Moreover, while Control 2 does have the highest tangent delta value at 20°C, its T_g is also much higher than any of the examples. This fact indicates that the low temperature performance of this formulation has been

30 compromised to achieve that high a tangent delta value. In fact, because the T_g is above 15°C, whereas the T_g of each of the examples is below 10°C, the adhesive or pressure sensitive properties of the formulation of Control 2 will be generally unacceptable for the great majority of

35 conventional applications. The examples all have T_g values comparable to Control 1, and all the examples show significant increases in tangent delta values at 20°C over Control 1. The pressure sensitive adhesives of the present

invention exhibit a single T_g value. This single T_g value is the result of the styrene isoprene di-block copolymer and the remaining elastomers being miscible with the tackifier. Accordingly, the examples possess wider temperature ranges than the controls and the improved tangent delta values indicate a substantial improvement in ease of conversion.

Table 1

	Control 1	Control 2	Example 1	Example 2	Example 3
Shellflex 371	15	-	-	5	10
Wingtack 10	-	15	-	-	-
LVSI-101	-	-	15	10	5
Piccotac 95	55	55	55	55	55
Kraton 1107	30	30	30	30	30
Irganox 1010	1	1	1	1	1
180° Peel to Stainless Steel	4.9	5.5	5.5	5.6	5.9
Shear Adhesion Failure Temperature (°F)	164	162	179	168	168
T_g (°C)	7.17	16.34	9.37	8.99	7.12
Tangent Delta at 20°C	1.069	2.404	1.179	1.213	1.173

Table 2 compares Example 17 from Sasaki et al, U.S. Patent No. 5,290,842, here labeled Control 3, to a formulation provided by the invention, indicated as Example 4. The difference between the two formulations is the replacement of half of the processing oil, Shellflex 371, with the styrene isoprene di-block copolymer LVSI-101. The DMA testing for each of the formulations shown in Table 2 was performed at 1% strain. Again, as can be seen, the 180° peel test and shear adhesion failure temperature tests yield much improved figures for Example 4 as compared to Control 3, indicating a substantial improvement in adhesive properties over a formulation described in the Sasaki

patent as having "excellent adhesive properties". Further, the tangent delta figures indicate much easier converting, again over a formulation described by Sasaki as exhibiting "excellent convertibility".

5

Table 2

10

15

	Control 3	Example 4
LVSI-101	-	12
Solprene 1205	21.2	21.2
Kraton D-1112	16.8	16.8
Escorez 1310 LC	38	38
Shellflex 371	24	12
Ethyl 330 ^a	0.7	0.7
Cyanox LTDP ^b	0.7	0.7
180° Peel to Stainless Steel	2.8	3.9
Shear Adhesion Failure Temperature (°F)	130	154
T _g (°C)	-7.12	-4.9
Tangent Delta at 20°C	0.556	0.696

^aphenolic antioxidant; ^bpropionic acid antioxidant.

Finally, the effect of other, non-styrene-
 20 containing low molecular weight polymers was compared, by comparing the formulation from Example 1 above to formulations using these polymers. In particular, low molecular weight isoprene rubber polymers LIR-30 and LIR-50, from Kuraray, through Arakawa, were selected for
 25 Controls 4 and 5. Here again, the DMA testing for each of the formulations shown in Table 3 was performed at 5% strain. Here the control formulations provide neither the peel adhesion nor the high temperature performance of a low molecular weight styrene-isoprene copolymer.

Table 3

	Control 4	Control 5	Example 1
LVSI-101	-	-	15
Piccotac 95	55	55	55
Kraton 1107	30	30	30
LIR-30	15	-	-
LIR-50	-	15	-
Irganox 1010	1	1	1
180° Peel to Stainless Steel	4.1	4.8	5.5
Shear Adhesion Failure Temperature (°F)	154	167	179
T _g (°C)	4.12	3.87	9.37
Tangent Delta at 20°C	1.124	0.928	1.179

While the adhesives hereinbefore described are effectively adapted to fulfill the aforesaid objects, it is to be understood that the invention is not intended to be limited to the specific preferred embodiments of pressure sensitive labeling adhesives set forth above. In particular without limitation, it should be understood that the formulations discussed herein may include additional oils, fillers, extenders, pigments, dyes, indicators, stabilizers, and other such ingredients as may be desired to achieve certain desirable or desired properties or avoid certain undesirable or undesired properties. Additionally, the formulations discussed herein may be free of organic solvents. The claimed invention, however, is to be taken as including all reasonable equivalents to the subject matter of the appended claims.

Claims:

1. A pressure sensitive adhesive, comprising:
an elastomer selected from the group consisting
of a styrene-isoprene-styrene block copolymer, a styrene-
isoprene-styrene/styrene-isoprene block copolymer, a
5 styrene-butadiene-styrene block copolymer, a styrene-
butadiene-styrene/styrene-butadiene block copolymer, a
styrene-butadiene block copolymer, a styrene-ethylene-
butylene-styrene block copolymer, a styrene-ethylene-
butylene-styrene/styrene-ethylene-butylene block copolymer
10 and a styrene-ethylene-propylene-styrene/styrene-ethylene-
propylene block copolymer, and mixtures thereof;
a tackifier; and
a styrene isoprene di-block copolymer.
2. A composition as recited in claim 1, wherein
the pressure sensitive adhesive has a single glass
transition temperature value.
3. A composition as recited in claim 2, wherein
the pressure sensitive adhesive has a tangent delta value
at 20°C which is increased by at least 5% compared to the
tangent delta value measured in the absence of the styrene
5 isoprene di-block copolymer, and wherein the glass
transition temperature is no greater than 15°C, and wherein
the shear adhesion failure temperature is not decreased
more than 5°F compared to the shear adhesion failure
temperature measured in the absence of the styrene isoprene
10 di-block copolymer.
4. A composition as recited in claim 2, wherein
the pressure sensitive adhesive has a tangent delta value
at 20°C which is increased by at least 10% compared to the
tangent delta value measured in the absence of the styrene
5 isoprene di-block copolymer, and wherein the glass
transition temperature is no greater than 15°C, and wherein
the shear adhesion failure temperature is not decreased
more than 5°F compared to the shear adhesion failure

temperature measured in the absence of the styrene isoprene
10. di-block copolymer.

5. A composition as recited in claim 1, wherein the styrene isoprene di-block copolymer has an absolute molecular weight of less than 70,000.

6. A composition as recited in any of claims 1 to 5, wherein the tackifier is present in an amount of about 5% to 70% by weight and the styrene isoprene di-block copolymer is present in an amount of about 3% to 40% by weight.

7. A composition as claimed in claims 1 to 6, wherein the composition further contains a plasticizer, the plasticizer being composed of a styrene isoprene di-block copolymer and a normally liquid tackifier.

8. A composition as claimed in claim 7, wherein the plasticizer is composed of a styrene isoprene di-block copolymer and a processing oil in a weight ratio of about 1:2 to 1:0.5.

9. A composition as recited in claim 7, wherein the elastomer is present in an amount of about 10% to 55% by weight, the tackifier is present in an amount of about 5% to 70% by weight, the plasticizer is present in an
5 amount of about 3% to 40% by weight, and the styrene isoprene di-block copolymer and processing oil is present in a ratio of about 1:2 to 1:0.5.

10. A laminate suitable for use as a label, comprising a face stock, a pressure sensitive adhesive layer, and a release liner, wherein the pressure sensitive adhesive layer consists essentially of the composition of claims 1 to 9.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US97/02582

A. CLASSIFICATION OF SUBJECT MATTER IPC(6) : B32B 7/12; C08L 57/02, 53/02, 93/04; C08K 5/01 US CL : Please See Extra Sheet. According to International Patent Classification (IPC) or to both national classification and IPC														
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) U.S. : 428/41.3, 41.8, 352, 354, 355; 524/ 271, 274, 484, 485, 486, 499, 505; 525/89, 98, 99 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)														
C. DOCUMENTS CONSIDERED TO BE RELEVANT														
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.												
X	US, A, 5,274,036 (KORPMAN et al.) 28 December 1993.	1-4												
---	See entire document.	-----												
Y		5												
X	US, A, 3,919,160 (LAKSHMANAN) 11 November 1975. See entire document.	1-5												
X	US, A, 3,972,848 (LAKSHMANAN) 03 August 1976. See entire document.	1-5												
Y	US, A, 5,290,842 (SASAKI et al.) 01 March 1994. See entire document.	1-5												
Y	US, A, 5,322,876 (SASAKI et al.) 21 June 1994. See entire document.	1-5												
<input type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family annex.														
<table border="0"> <tr> <td>* Special categories of cited documents:</td> <td>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</td> </tr> <tr> <td>"A" document defining the general state of the art which is not considered to be of particular relevance</td> <td>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</td> </tr> <tr> <td>"E" earlier document published on or after the international filing date</td> <td>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</td> </tr> <tr> <td>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</td> <td>"&" document member of the same patent family</td> </tr> <tr> <td>"O" document referring to an oral disclosure, use, exhibition or other means</td> <td></td> </tr> <tr> <td>"P" document published prior to the international filing date but later than the priority date claimed</td> <td></td> </tr> </table>			* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention	"A" document defining the general state of the art which is not considered to be of particular relevance	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone	"E" earlier document published on or after the international filing date	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art	"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&" document member of the same patent family	"O" document referring to an oral disclosure, use, exhibition or other means		"P" document published prior to the international filing date but later than the priority date claimed	
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Date of the actual completion of the international search		Date of mailing of the international search report												
09 APRIL 1997		11 JUL 1997												
Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703) 305-3230		Authorized officer <i>IW for</i> PETER SZEKELY Telephone No. (703) 308-1235												

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US97/02582**Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)**

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☒ Claims Nos.: 6-10
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US97/02582

A. CLASSIFICATION OF SUBJECT MATTER:

US CL :

428/41.3, 41.8, 353, 354, 355; 524/ 271, 274, 484, 485, 486, 499, 505; 525/89, 98, 99